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NEWS OF SCHOOLS OF HIGHER EDUCATION, PHYSICS  
(SELECTED ARTICLES)



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NEWS OF SCHOOLS OF HIGHER EDUCATION. PHYSICS  
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## ON THE NATURE OF DIELECTRIC LOSSES IN MICA

I. G. Vorozhtsova

### Introduction

The dielectric properties of mica have been studied fairly thoroughly between  $-180^{\circ}$  and  $+300^{\circ}\text{C}$  and in the frequency range from 50 cps to 10 Mc [1, 2, 3, 4]. The presence of two temperature [1, 2] and two frequency maxima for the loss angle has been established in phlogopite [3] and muscovite [4], the latter roasted at  $600^{\circ}\text{C}$ . The low-temperature maximum for  $\tan \delta$  has the usual relaxation character, while the high-temperature maximum behaves unusually, viz., with an increase in frequency it is displaced toward the lower temperatures and is accompanied by a decrease in permittivity with an increase in temperature.

The nature of the high-temperature loss-angle maximum has still not been clarified, and the kind of relaxers causing the low-temperature maximum of  $\tan \delta$  is also not clear. The theories in the literature which deal with these questions are often contradictory.

For example, Van Keymeulen [3] considers that the low-temperature loss-angle maximum in phlogopite is due to the orientation of pseudodipoles created by defects in the atomic valence structure (Breckenridge relaxation phenomena [6]). M. S. Metsik [5] assumes

a description-ohmic interpretation of the dielectric losses in mica. Finally, K. A. Vodop'yanov and his pupils [1, 2, 4, 7] connect the dielectric losses in mica with the possibility of orientation of crystal-water molecules.

In one of the author's previous papers [4] an assumption was made connecting the two temperature maxima of the loss angle in mica with the respective relaxation polarizations of the two groups of water contained in it (semibound and constitutional).

In order to test this assumption, it is interesting to study the dielectric properties of mica under reduced air pressure. In view of the weak attachment of semibound water (5b), we may expect that a change in the external conditions (temperature and pressure) will have an effect on the nature of the temperature dependence of  $\tan \delta$  and  $\epsilon$ , primarily on the shape of the low-temperature maximum.

#### Experimental Results and Discussion

Muscovite roasted at 600°C (M-600) was chosen for the study since the less-angle maxima are most clearly expressed in this material.

The temperature-frequency dependence of  $\tan \delta$  and  $\epsilon$  in M-600 were studied in the frequency range from 20 to  $10^5$  cps from -80 to +250°C under conditions of atmospheric pressure and under reduced air pressure ( $10^{-4}$  mm Hg).

The specimens were disc-shaped and 0.25 mm thick; the diameter of the electrodes was 22 mm. Before measurement the specimens were rinsed with benzene and dried in a vacuum oven at 150°C.

The frequency dependences of  $\tan \delta$  and  $\epsilon$  in M-600 for various temperatures are presented in Figs. 1 and 2. It is apparent from Fig. 1 that at temperatures below 100°C the dielectric losses in heat-treated muscovite have all the indications of a relaxation

mechanism:

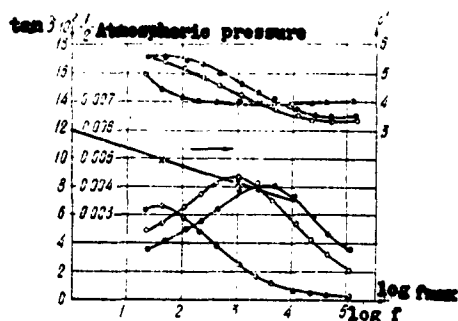


Fig. 1. Frequency dependences of  $\tan \delta$  and  $\epsilon$  in muscovite roasted at 600°C (M-600). ● at  $T = -70^\circ\text{C}$ ; ○ at  $T = -36^\circ\text{C}$ ; ● at  $T = +20^\circ\text{C}$  under conditions of atmospheric pressure; X  $\log f_{\max}$  as a function of  $1/T$ .

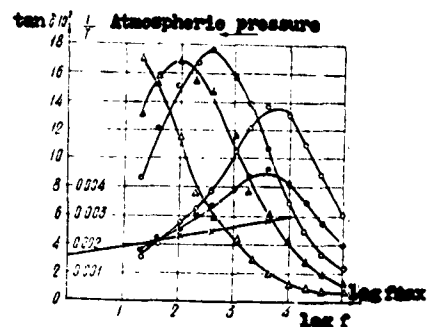


Fig. 2. Frequency dependences of  $\tan \delta$  in M-600. ● at  $T = 40^\circ\text{C}$ ; ○ at  $T = 94^\circ\text{C}$ ; ⊙ at  $T = 140^\circ\text{C}$ ; ▲ at  $T = 180^\circ\text{C}$ ; Δ at  $T = 240^\circ\text{C}$  under atmospheric pressure; X  $\log f_{\max}$  as a function of  $1/T$ .

- 1) the frequency maximum of the dielectric loss angle is displaced toward the higher frequencies, as the temperature increases;
- 2) the permittivity in the region of the maximum of  $\tan \delta$  decreases, as the frequency increases;
- 3)  $\log f_{\max}$  increases in proportion to  $T$ , where  $f_{\max}$  is the maximum loss-angle frequency and  $T$  is the absolute temperature.

From the results given in Fig. 1 we determined the activation energy of the relaxers, which proved to be equal to 0.39 eV.

Above  $+94^\circ\text{C}$ , as can be seen from Fig. 2, the following is observed:

- 1) The displacement of the loss-angle maximum reverses direction, that is, with an increase in temperature the maximum of  $\tan \delta$  begins to be displaced toward the lower frequencies;
- 2) the directly proportional dependence of  $\log f_{\max}$  on  $T$  changes to an inversely proportional dependence.

The activation energy, which is determined from the displacement of the  $\tan \delta$  maximum, decreases with an increase in temperature,



assuming negative values above +94°C (Table 1).

The temperature-frequency dependences of the same muscovite specimen in a vacuum under a pressure of  $10^{-4}$  mm Hg are given in Fig. 3. The arrow indicates the motion of the frequency maximum of the loss angle as the pressure increases. It is apparent from the figure that the reversal of direction of the frequency maximum under reduced pressure begins at 40°C, that is, at a temperature 50-60°C lower than under atmospheric pressure.

Thus the displacement of the  $\tan \delta$  maximum reverses direction under reduced pressure, as well as under atmospheric pressure, in heat-treated muscovite.

The temperature dependence of the loss angle in muscovite roasted at 600°C, as can be seen from Fig. 4, is of a complicated nature, assuming the shape of a curve with two maxima. The permittivity in the region of the low-temperature maximum of  $\tan \delta$  increases with temperature, while in the region of the high-temperature maximum it decreases.

When the pressure of the air surrounding the specimen is reduced to  $10^{-7}$  mm Hg,\*\* the nature of the temperature dependence of the loss angle and of the permittivity remains unchanged, but the high-temperature maximum of  $\tan \delta$  is displaced toward lower temperatures by 100°C.

This result is unexpected, since it was assumed that the high-temperature maximum was caused by the orientation of constitutional-water molecules, which are attached in the crystal more strongly

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\*\* Trans. Note:  $10^{-7}$  mm Hg...Numeral omitted from exponent in original.

than the molecules of semibonded water. Therefore a decrease in the air pressure should have primarily affected the low-temperature maximum of the loss angle in mica.

This set of circumstances, together with the fact that at any temperature, there is only one frequency maximum of the loss angle in the broad range of frequencies ( $20 \cdot 10^{-5}$  cps) corresponding to the two temperature maxima of  $\tan \delta$  leads us to think that under the observed conditions there is only one relaxation mechanism in roasted muscovite instead of the expected two. It is likely that under the given conditions only the semibound water molecules are oriented.

The question arises: what is the mechanism causing the reversal in the direction of the displacement of the frequency maximum, i.e., the observed breakdown in the relaxation nature of the dielectric losses?

We shall assume that this phenomenon is due to structural changes occurring in muscovite during its heating, more precisely speaking, to the change in the number of relaxers accompanying these changes.

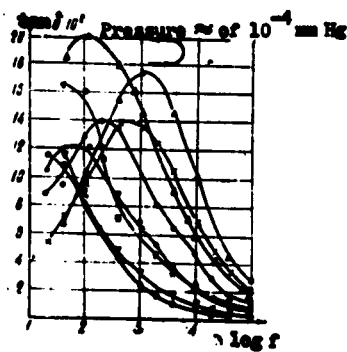


Fig. 3. Frequency dependences of  $\tan \delta$  in M-600. ● at  $T = -48^\circ\text{C}$ ; ○ at  $T = -30^\circ\text{C}$ ; ● at  $T = -9^\circ\text{C}$ ; X at  $T = 6^\circ\text{C}$ ; ▲ at  $T = 40^\circ\text{C}$ ; Δ at  $T = 66^\circ\text{C}$ ; \* at  $T = 110^\circ\text{C}$ ; V at  $T = 140^\circ\text{C}$ ; air pressure  $\approx 10^{-4}$  mm Hg.

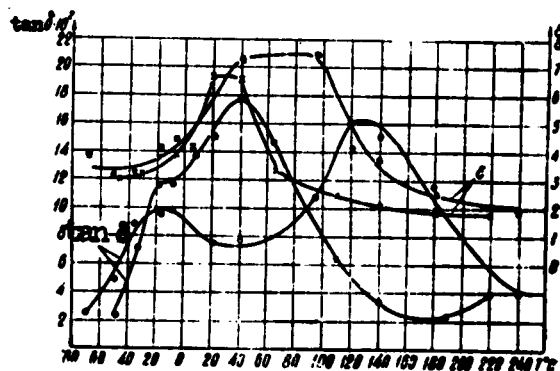


Fig. 4. Frequency dependences of  $\epsilon$  and  $\tan \delta$  in M-600 at a frequency of  $10^5$  cps at atmospheric pressure (\*, O) and in a vacuum (x, ●).

It is characteristic of mica that there are certain temperature ranges in which a sudden change occurs in the physical and chemical properties of the mica (thermal conductivity, linear expansion, swelling), which is accompanied by thermographic and heat-weight effects, as well as by a change in structure, which is recorded on Laue crystallograms as mosaic reflexes [8] and on Debye crystallograms as a broadening of the lines [9].

The nature of the structural changes causing these effects is still undisclosed. Wood [9] and other authors assume that the formation of mosaic structures in mica occurs not as a result of the evolution of gaseous products (steam) during heating, but as a result of a change in the atomic structure. The reversibility of the processes occurring in mica during heating and the presence of a critical temperature lead to this way of thinking. Metsik and Zhidikhanov [10] explain the sudden swelling of mica in a certain temperature range by the transformation of water into steam in closed microcavities.

In any case, structural changes do occur in mica between 20 and 300°C, and cannot fail to affect the dielectric losses in it.

Above all, the dehydration of the semi-bound water from the crystal, which accompanies these changes, must have an effect. In fact, if it has been accurately assumed that in a given range of temperatures and frequencies the dielectric losses in mica are determined by relaxation polarization due to orientation of semi bound water molecules, then the removal of this water, i.e., a decrease in the number of relaxers, cannot fail to affect the nature of the temperature frequency dependences of  $\tan \delta$  and  $\epsilon$  in roasted muscovite.\*

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\* Note that these changes are not observed in nonroasted muscovite [8] and also that there are no maxima on the temperature-frequency curves of  $\tan \delta$  in nonroasted muscovite [4].

At the present time theoretical papers [11, 12] describing the polarization in a dielectric with a variable number of relaxers are known. For example, Volokobinskiy [11] considered a case of an increase in the number of relaxers and explained certain features of the dielectric losses in glasses. I. Ts. Lyast [12] proposed a theory of relaxation polarization of crystal hydrates. By solving a kinetic equation he found an expression for the complex permittivity. In Lyast's theory the relaxation time  $\tau$  is expressed in terms of the number of relaxers, so that as the number of the latter decreases  $\tau$  will increase, i.e., the loss-angle maximum should occur at lower frequencies. Accordingly, the activation energy, which is determined from the displacement of the frequency maximum of  $\tan \delta$ , should decrease as the temperature increases, which is what is observed in the experiment (Table 1).

Thus certain deviations of the experimental results in mica from the general theory of dielectric losses [13], such as: 1) reversal of the direction of the displacement of the frequency maximum of  $\tan \delta$ , when the temperature is increased, 2) a decrease in permittivity accompanying a rise in temperature in the region of the high-temperature maximum of  $\tan \delta$  may be explained, on the basis of Lyast's assumption, by a decrease in the number of relaxers in the crystal.

#### Taking Into Account the Set of Relaxation Times

As is known, the frequency maxima of  $\tan \delta$  which characterize relaxation dielectric losses, when obtained by experiment, are lower in absolute value and broader than the theoretical. This circumstance is explained by the presence of a set of relaxation times in a real dielectric.

In order to estimate the discrepancy between theory and experiment,

K. Cole and R. Cole suggested representing the experimental results in the form of curves of  $\epsilon''$  versus  $\epsilon'$ , where  $\epsilon'' = \epsilon' \tan \delta$ . In this representation the trajectory of Debye's equations

$$\begin{aligned}\epsilon' &= \epsilon_\infty + (\epsilon_0 - \epsilon_\infty) / (1 + \omega^2 \Theta^2); \\ \epsilon'' &= (\epsilon_0 - \epsilon_\infty) \omega \Theta / (1 + \omega^2 \Theta^2)\end{aligned}\quad (1)$$

is a semicircle with its center on the real axis  $\epsilon'$  and the points of intersection with this axis being  $\epsilon_0$  and  $\epsilon_\infty$ .

An analysis of a huge amount of experimental material revealed that, instead of the theoretical semicircle, an arc with its center lying below the axis is obtained by experiment. The angle  $\alpha$  between the  $\epsilon'$  axis and the arc radius drawn through the point  $\epsilon_\infty$  has been chosen as the measure of the deviation of the experiment from the theory.

Analytically, the experimental arc is expressed by the equations

$$\begin{aligned}\epsilon' &= \frac{\epsilon_\infty + (\epsilon_0 - \epsilon_\infty) \left[ 1 + (\omega \Theta)^{-1} \sin \frac{1}{2} \alpha \pi \right]}{1 + 2(\omega \Theta)^{1-\alpha} \sin \frac{1}{2} \alpha \pi + (\omega \Theta)^{2(1-\alpha)}}; \\ \epsilon'' &= \frac{(\epsilon_0 - \epsilon_\infty) (\omega \Theta)^{1-\alpha} \cos \frac{1}{2} \alpha \pi}{1 + 2(\omega \Theta)^{1-\alpha} \sin \frac{1}{2} \alpha \pi + (\omega \Theta)^{2(1-\alpha)}},\end{aligned}\quad (2)$$

which differ from Debye's only in the parameter  $\alpha$ .

Applying a method [15] for finding the distribution function of the relaxation times from the dependence  $\epsilon'' = f(\omega)$  known from experiment and using Eqs. (2) in this capacity, Cole and Cole found this function in the form

$$F(s) = \frac{1}{2\pi} \frac{\sin \pi \alpha}{\cosh[(1-\alpha)s - \cos \pi \alpha]}, \text{ where } s = \ln \left( \frac{\tau}{\tau_m} \right). \quad (3)$$

Thus the parameter  $\alpha$  characterizes the packing density of the relaxation times about their most probable value. Depicting the experimental results by the method of circle diagrams is very convenient, since it enables us to judge the presence of a set of relaxation times, and also to calculate the maximum value of  $\tan \delta$  more accurately, using a formula found from Eqs. (2) [16]:

$$\tan \delta_{\max} = \frac{(z_0 - z_{\infty}) \cos \frac{1}{2} \alpha \pi \sqrt{\frac{z_0}{z_{\infty}}}}{2z_0 + \sqrt{\frac{z_0}{z_{\infty}} (z_0 - z_{\infty}) \sin \frac{1}{2} \alpha \pi}} \quad (4)$$

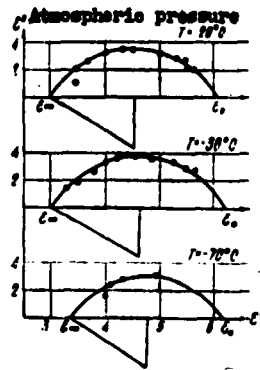


Fig. 5. Circle diagrams for M-600 at low temperature.

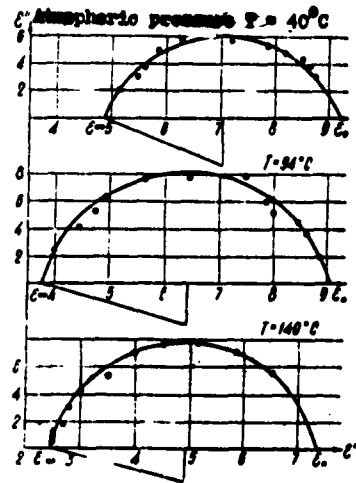


Fig. 6. Circle diagrams for M-600 at high temperatures.

Circle diagrams for muscovite roasted at 600°C are shown in Figs. 5, 6, and 7. As can be seen from the figures, the circle diagrams of the investigated mica have the form of arcs, and the parameter  $\alpha$  is fairly high (up to 0.53 radians), which attests to the appreciable width of the set of relaxation times in muscovite.

It is interesting to note that for an increase in temperature (above 90°C), as well as for a decrease in pressure, a decrease in the parameter  $\alpha$  is observed (Figs. 5 and 6). This fact attests to a contraction of the set of relaxation times under conditions favorable

TABLE 1 \*

Pressure	T °C	log f <sub>max</sub>	Activation energy in eV
	-70	1.6	—
	-56	3.0	0.30
	20	3.4	0.10
	40	3.6	0.18
	94	3.75	0.00
	140	2.5	-0.80
	180	2.0	-0.50
	—	—	—
	-19	2.3	0.55
	-6	2.85	0.20
	-40	3.1	0.14
	-66	2.0	-0.88

at atmospheric pressure

at 10<sup>-4</sup> mm Hg.

TABLE 2

Temperature, °C	Report- initial	According to Neuge- bauer	Report- initial	According to Neuge- bauer	Report- initial	According to Neuge- bauer	Report- initial	According to Neuge- bauer	Report- initial	According to Neuge- bauer	Report- initial	According to Neuge- bauer	Report- initial	According to Neuge- bauer	Report- initial	According to Neuge- bauer	Report- initial	According to Neuge- bauer
Temperature, °C	-70	—	-36	—	20	—	94	—	140	—	140	—	140	—	140	—	140	—
tan δ <sub>max</sub> at atmospheric pressure	0.067	0.33	0.083	0.086	0.37	0.056	0.08	0.36	0.075	0.130	0.450	0.170	0.57	0.16	0.16	0.16	0.16	0.16
Temperature, °C	-30	—	-19	—	6	—	40	—	66	—	66	—	66	—	66	—	66	—
tan δ <sub>max</sub> at a pressure of 10 <sup>-4</sup> mm Hg.	0.12	0.43	0.19	0.14	0.44	0.2	0.14	0.43	0.2	0.18	0.47	0.27	0.2	0.82	0.82	0.82	0.82	0.82

\* The active energy was determined for two neighboring temperatures.

to intense desorption of water from the crystal.

The results of calculating  $\tan \delta$  from Debye's formula  $\tan \delta_{\max} = (\epsilon_0 - \epsilon_\infty) / 2\sqrt{\epsilon_0 - \epsilon_\infty}$  and the Coles' formula (4), are given in Table 2 and attest to the fairly good agreement between the experimental results and the Coles' formula.

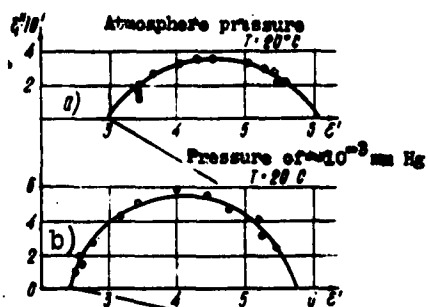


Fig. 7. Circle diagrams for M-600 at various air pressures.

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## ON THE MECHANISM OF THE ELECTRIC BREAKDOWN OF SOLID DIELECTRICS

G. A. Vorob'yev

Many laws of the electric breakdown of solid dielectrics and air are similar. However, there are no experimental data which would directly indicate the breakdown mechanism of solid dielectrics. Modern theories of the electric breakdown of solid dielectrics do not consider the development of discharge, and therefore do not give a direction for experimental study of the mechanism of breakdown. An examination of collision ionization in a sample of rock salt 0.1 mm thick indicates the size of the electronic avalanche is  $10^{30}$ , which is unreal. A factor which limits the growth of an electronic avalanche, as calculations show, is the space charge formed as a result of collision ionization. Only in a sample with a thickness of several microns, where the space charge is small, can collision ionization be observed in pure form. In this case hardening of the dielectric, an increase in the delay time, and an exponential rise in current must be observed. Experiments conducted [1] confirmed these assumptions.

The electric breakdown of solid dielectrics has been studied for over thirty years. There are a considerable number of theories [1, 2, 3, 4] which explain differently the mechanism of electric breakdown of solid dielectrics. The main mechanisms of breakdown given in these theories are the following: rupture of the crystal lattice by the

electrostatic forces of the field, collision ionization by ions, electrostatic ionization (tunnel effect, thermal ionization facilitated by the field, electron emission from the cathode), collision ionization by electrons. Experimental data and the results of calculations show a low probability for these mechanisms, with the exception of the last. Therefore, it is now thought that collision ionization by electrons is responsible for breakdown of solid dielectrics.

However, there was no experimental proof for this. The Hippel-Callen and Frelich theories of collision ionization in solid dielectrics pertain to a single act of ionization. For direction in experimental studies it is necessary to have a theory which considers not only a single act of ionization, but also the development of discharge in space and time. There is no such theory at the present time.

Some questions on the development of discharge in solid dielectrics are considered in this article, assuming that discharge starts from collision ionization.

Rock salt (NaCl) will be used as the example for the study. Let the ionization potential  $U_1 = 9.6$  v and the electric strength

$$E_s = 1.5 \cdot 10^6 \text{ v/cm.}$$

The minimum path length over which an electron can gain energy sufficient for ionization is

$$\lambda_{\min} = \frac{U_1}{E_s} = 6.4 \cdot 10^{-6} \text{ cm.}$$

But the actual of  $\lambda$  must exceed  $\lambda_{\min}$ , owing to the loss of energy by an electron as it is accelerated in the crystal lattice.

It has been shown [9] that in air at atmospheric pressure approximately 1/124 of the energy of the electron accumulated in the electrical field is expended by collision ionization. The remaining electron

energy is spent in warm-up of the air and in radiation. If this were valid for rock salt, then

$$\lambda = \lambda_{\min} \cdot 124 \approx 8 \cdot 10^{-4} \text{ cm.}$$

However, owing to the low probability of excitation of points of the crystal lattice, the energy losses by an electron in the crystal are considerably less than in a gas [4]. Therefore, the actual value of  $\lambda$  in rock salt must be between  $6.4$  and  $8 \cdot 10^{-4}$  cm.

According to calculations by V. A. Chuyenkov [6], the magnitude of  $\lambda$  in an NaCl crystal is  $10^{-4}$  cm. This value can be taken for later calculations. Then, in a rock salt sample, whose thickness in tests is usually on the order of tenths of millimeters, the number of electron in the avalanche reaching the anode is  $n_a = 2^{d/\lambda} = 2^{100} \approx 10^{30}$ , if  $d = 0.1$  mm.

This quantity of electrons, if the valence electrons and those in the inner shells are counted, is contained in about 15 tons of rock salt. Hence it is clear that the development of an electronic avalanche in solid dielectrics, besides collision ionization, is affected also by other factors which slow down its development. A positive space charge should be considered the main factor arising as a result of collision ionization. It is known that a space charge arising as a result of collision ionization strongly affects the development of discharge in gases. It has been shown [6] that it can affect the development of discharge even in a solid dielectric.

Figure 1 shows a positive space charge and an electronic avalanche at a certain stage of development. For simplicity, let us assume that the boundary between both types of charge is a spherical surface. The highest field strength created by both charges will be at the point of tangency of the spherical surfaces (point a in Fig. 1) and will

equal  $E = E_1 + E_2$ . This field strength is opposite in direction to the applied one  $E_p$ .

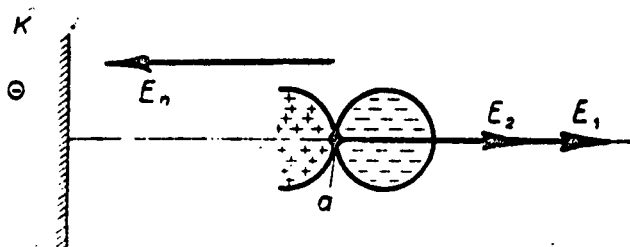


Fig. 1. Positive space charge and electronic avalanche at a certain stage of development.  $K$  is the cathode and  $a$  is the point of tangency of the hemispheres.

$E_1$  and  $E_2$  can be evaluated:  $E_1 = \frac{q_1}{4\pi\epsilon r^2}$  and  $E_2 = \frac{q_2}{4\pi\epsilon r^2}$ , where  $q_1$  and  $q_2$  are the electron and ion charges respectively;  $q_1 = 2^{x/\lambda} \cdot e$ , where  $x$  is the path traveled by the electronic avalanche. At each place at which ionization occurs, the number of ions is equal to the number newly formed free electrons and, therefore, is less than the total number of electrons by a factor of 2. Thus

$$q_2 = \frac{q_1}{2}.$$

The radius  $r$  of the head of the electronic avalanche, which is dependent upon transverse electron diffusion, is determined, according to Seitz [7], by the expression  $r = \sqrt{Dt}$ , where  $D$  is the diffusion factor, which, according to Seitz [7], can be taken as  $1 \text{ cm}^2/\text{sec}$ . The time of development of the avalanche  $t = \frac{x}{kE_p}$ , where  $k$  is the electronic mobility. Let us take  $k = 1 \text{ cm}^2/\text{v} \cdot \text{sec}$  [7].

The results of the calculation are given in Table 1.

From Table 1 it is apparent that after 13 ionizations, if the number of electrons grew according to the law of collision ionization,

$E_1 + E_2$  already exceeds the strength of the applied field  $E_p$ . Just as  $E_1 + E_2$  becomes comparable with  $E_p$ , a part of the electrons will be delayed by the positive space charge, and the greater it is, the greater  $E_1 + E_2$ . This will retard the growth of the electronic avalanche

TABLE 1

$\frac{N}{L}$	$t_i$ $10^{-10}$ sec	$r_i$ $10^{-5}$ cm	$n_e$	$E_1$ v/cm	$E_2$ v/cm	$E_1 + E_2$ v/cm
5	3,3	1,8	32	$1,38 \cdot 10^4$	$0,69 \cdot 10^4$	$2,17 \cdot 10^4$
8	5,3	2,3	256	$7 \cdot 10^4$	$3,5 \cdot 10^4$	$1,05 \cdot 10^5$
9	6	2,44	512	$1,24 \cdot 10^5$	$0,62 \cdot 10^5$	$1,86 \cdot 10^5$
10	6,66	2,58	$\sim 10^3$	$2,16 \cdot 10^5$	$1,08 \cdot 10^5$	$3,24 \cdot 10^5$
12	8	2,83	$\sim 4 \cdot 10^4$	$7,2 \cdot 10^5$	$3,6 \cdot 10^5$	$1,08 \cdot 10^6$
13	8,67	2,94	$\sim 8 \cdot 10^4$	$1,33 \cdot 10^6$	$0,66 \cdot 10^6$	$2 \cdot 10^6$

Collision ionization will occur in pure form at thickness  $\underline{d}$  when  $E_1 + E_2 \ll E_p$ . If we take  $E_1 + E_2 = 0.1E_p$ , then collision ionization can be observed in undistorted form when  $\underline{d}$  does not exceed  $d_0 = 9\mu$  (Table 1), if  $\lambda = 10^{-4}$  cm.

If  $d < d_0$ , then average size of the electronic avalanche  $n_e = 2^{d/\lambda}$  can be insufficient for formation of a conducting path between electrons, and, therefore, this can cause breakdown of the dielectric.

Two possible mechanisms of breakdown can be proposed for this case:

1. A conducting path between electrons is created by a single avalanche, but the probability of the appearance of such an avalanche is low. The delay time of a discharge in this case is the statistical delay time, and it has been examined by Seitz [7].

2. A conducting path between electrons is created by the

accumulation of conducting plasma from each avalanche. The delay time of discharge in this case is the discharge formation time.

In both cases the discharge time is a function of the output frequency of electrons  $\nu_0$  from the cathode, and can be significant if  $\nu_0$  is not too high.

In a thin layer of dielectric, as compared with a thicker one, one should also expect an increase in electric strength, which follows from collision-ionization theory, and also an increase in the current in the dielectric according to the law  $i = i_0 2^{d/\lambda}$ , where  $i_0$  is the emission current from the cathode.

According to the instructions of Professor A. A. Vorob'yev and ours, V. A. Kostrygin studied the delay time of discharge and electric strength in rock salt with a thickness of  $10^{-4}$  to  $10^{-3}$  cm [8]. He discovered an increase in electric strength with an increase in thickness, and a considerable increase in delay time.

According to V. A. Kostrygin's data [8], in rock salt with a thickness of  $10^{-4}$  to  $10^{-3}$  cm, the delay time is on the order of  $10^{-8}$  sec; according to K. K. Sonchik's data [9], in rock salt with a thickness of 0.12 mm, the delay time of discharge is on the order of  $10^{-8}$  sec. The qualitative experimental confirmation of the assumptions made in the present work can be considered an indication that breakdown of solid dielectrics begins with collision ionization.

The author thanks Professor Doctor A. A. Vorob'yev for scientific guidance.

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## THE INFLUENCE OF GAMMA RADIATION ON DIELECTRIC LOSSES IN MICA

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Mineralogical forms of mica — muscovite and phlogopite — have vast engineering applications in the initial state as well as in the form of additives to various electrical insulator products used in the atomic industry. However, there are almost no data in the literature about the change in the electrical characteristics of muscovite and phlogopite under the influence of radiation. The effect of radiation on the electrical conductivity of mica has been studied chiefly [1].

Because many mica parts must work not only under conditions of heightened radiation, but also at high temperatures, it is necessary to study the effect of radiation on the dielectric properties of mica subjected to preliminary heat treatment. In addition, the study of dielectric losses in irradiated mica is also of theoretical interest, with the aim of explaining processes caused by irradiation.

The present work is a study of the effect of gamma radiation on the nature of the curve of temperature versus  $\tan \delta$  and  $\epsilon$  for muscovite over the temperature range from 20 to 300°C and the frequency range from 50 to  $10^4$  cps. Unroasted and heat-treated muscovite was studied.

Heat treatment of the mica consisted of roasting samples at 600 and 800°C for 4 hours. Irradiation was carried out at room temperature with a dose of 50,000 r and dose rates of 400 r/min and 1000 r/min. The radiation source was a betatron with a gamma-quantum energy of up to 15 Mev.

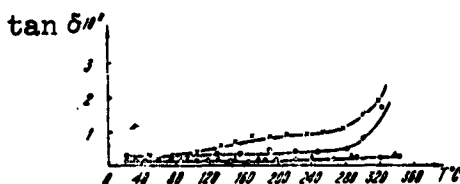


Fig. 1. Temperature versus  $\tan \delta$  for unroasted muscovite:  $\Delta, \times$ ) at 50 cps and  $\circ, \bullet$ ) at 10<sup>4</sup> cps;  $\Delta, \circ$ ) before irradiation;  $\times, \bullet$ ) after irradiation at a dose rate of 400 r/min, with a cumulative dose of 50,000 r.

Figure 1 shows temperature dependences of the loss angle in unroasted muscovite at frequencies of 50 and 10<sup>4</sup> cps before and after irradiation at a dose rate of 400 r/min. It is apparent from this figure that in muscovite there is an increase in  $\tan \delta$  for the irradiated sample over that of the unirradiated one. This difference increases with an increase in frequency.

After roasting at 600°C, the structure of the muscovite disintegrates somewhat [2,3], which leads to a change in the nature of the curve of temperature versus  $\tan \delta$ . In fact, the curve of the dependence of the loss angle in muscovite roasted at 600°C (M-600), as is apparent from Figs. 2 and 3, passes through a maximum in the temperature range from 80 to 160°C at 10<sup>4</sup> cps, and from 160 to 280°C at 50 cps. The dielectric constant over the temperature range of maximum  $\tan \delta$

decreases with an increase in temperature (Fig. 4)\*.

From a comparison of Figs. 2 and 3, it is apparent that irradiation acts on M-600 differently, depending upon the dose rate of gamma radiation.

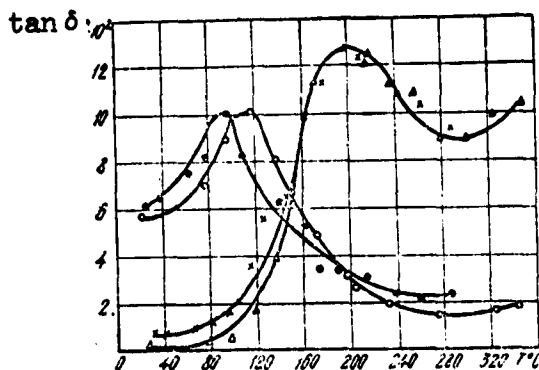


Fig. 2. Temperature versus  $\tan \delta$  for M-600:  $\Delta, \times$ ) at 50 cps and  $\circ, \bullet$ ) at  $10^4$  cps;  $\Delta, \circ$ ) before irradiation;  $\times, \bullet$ ) after irradiation with a dose rate of 400 r/min and a cumulative dose of 50,000 r.

After irradiation at a dose rate of 400 r/min, the nature of the curve of temperature versus  $\tan \delta$  does not change; only the maximum of the loss angle at a frequency  $10^4$  cps shifts to the lower frequency side by  $20^\circ C$ .

A similar result was obtained by us with phlogopite with irradiation under these same conditions [4].

An entirely different effect is observed, as is apparent from Fig. 3, for M-600 after irradiation at a dose rate of 1000 r/min:

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\*In order not to overload the figure, the results of measurements are given only for  $10^4$  cps.

irradiation leads to almost complete disappearance of the maximum of the loss angle in the sample, and to an increase in the high-temperature branch of the curve of  $\tan \delta$  versus temperature, especially great at the lower frequency.

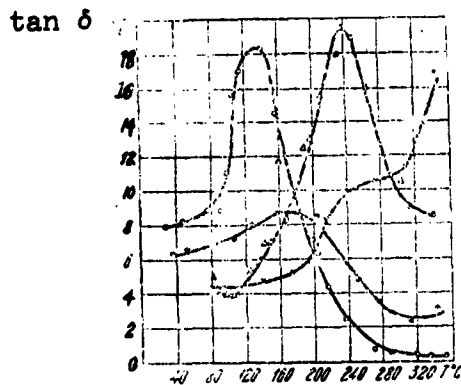


Fig. 3. Temperature versus  $\tan \delta$  for M-600:  $\Delta, \times$ ) at 50 cps and  $O, \bullet$ ) at  $10^4$  cps;  $\Delta, O$ ) before irradiation;  $\times, \bullet$ ) after irradiation at a dose rate of 1000 r/min with a cumulative dose of 50,000 r.

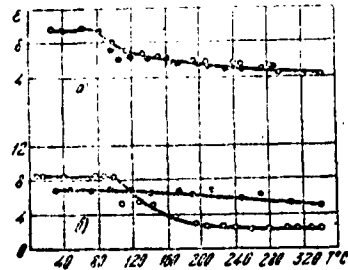


Fig. 4. Temperature versus  $\epsilon$  for M-600 at a frequency of  $10^4$  cps: a) before ( $O$ ) and after ( $\bullet$ ) irradiation at a dose rate of 400 r/min; b) before ( $O$ ) and after ( $\bullet$ ) irradiation at a dose rate of 1000 r/min. 50,000 r dose.

Figure 4 shows temperature versus dielectric constant for M-600 before and after irradiation. From Fig. 4a it is apparent that irradiation with a dose rate of 400 r/min does not affect the absolute value of  $\tan \delta$  over the entire temperature range from 20 to 300°C\*. After irradiation at a rate of 1000 r/min, the bend in the curve of the dielectric constant versus temperature disappears, due to the fact that the absolute value of  $\epsilon$  over the temperature range of maximum  $\tan \delta$  decreases, while at higher temperatures it increases in comparison with the value of  $\epsilon$  for an unirradiated sample.

Thus, in muscovite which has been roasted at 600°C, the dielectric properties are strongly dependent upon the dose rate. There seems to be a critical dose rate, above which irradiation leads to a decrease in the absolute value of  $\tan \delta_{\max}$  and to a change in the nature of the dependence of the loss angle upon temperature.

In order to explain these results, it is first of all necessary to study the nature of the observed maximum of the loss angle.

On the basis of a number of investigations [5, 6, 7], we assume that dielectric losses in mica are dependent upon the orientation of water molecules in the crystal. At the same time, certain deviations of experimental results from theory (a shift of the frequency maximum of  $\tan \delta$  with a rise in temperature in a direction opposite to that required by theory, the decrease in  $\epsilon$  over the temperature range of the maximum of the loss angle, etc.) are explained by us in previous works [6, 7] by the effect of dehydration of water molecules from the crystal

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\*At a frequency of 50 cps, the result of irradiation on the dielectric constant is similar to that at 10<sup>4</sup> cps.

on relaxation in the mica\*.

If we allow the possibility of decomposition of the semi-bound water in the mica into hydrogen and hydrogen peroxide by irradiation, as is well-known from numerous works on free water [9], then the obtained results can be explained as follows.

Firstly, the shifting of the maximum of  $\tan \delta$  to the low-temperature side after irradiation of M-600 at a dose rate of 400 r/min is explained by the decrease in the number of relaxation agents as a result of radiolysis of the water.

In fact, as I. Ts. Lyast showed in his work [8], an increase in relaxation time  $\tau$  with a decrease in the number of relaxation agents is characteristic of crystalline hydrates. But if this is so, then in order to fulfill the condition  $\omega\tau = 1$ , the temperature for the maximum value of the loss angle for a constant frequency  $\omega$  must be lowered.

Secondly, the decrease in the absolute value of the dielectric constant and the maximum value of the loss angle in M-600 after irradiation at a rate of 1000 r/min can be explained by the intensive decomposition of water with irradiation at a given dose rate, and, therefore, by the removal of a considerable amount of the relaxation agents.

The critical nature of the effect of the dose rate on dielectric losses in M-600 is analogous to the presence of a critical temperature for dehydration of water from mica. It is known that with a decrease

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\*It is assumed [6] that in the frequency range from 20 to  $10^5$  cps and in the temperature range from  $-80$  to  $300^\circ\text{C}$ , relaxation polarization occurs in M-600, which is dependent upon the orientation of molecules of the semi-bound water. The second water group (constitution) is bound more strongly, and the polarization connected with it will probably be observed at higher temperatures.

M. S. Metsik and R. A. Zhidikhanov have given a model of the distribution of semi-bound water in a mica crystal [8].

in the concentration of the water of crystallization in mica, the loss angle increases at first, and then upon reaching some concentration maximum it again decreases [10, 11].

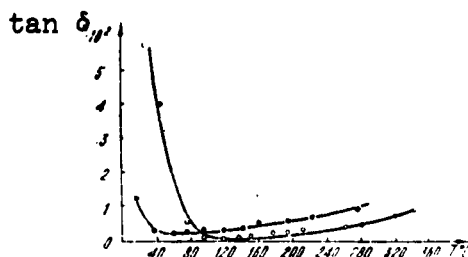


Fig. 5. Temperature versus  $\tan \delta$  for M-600 at a frequency of  $10^4$  cps: O) before irradiation; ●) after irradiation at a dose rate of 400 r/min with a cumulative dose of 50,000 r.

The result of irradiation of muscovite which was roasted at 800°C (M-800) (shown in Fig. 5) does not contradict the above explanation. The decrease in the absolute value of the loss angle over the temperature range from 20 to 80°C after irradiation of M-800 is due to the drying of the sample as a result of the radiolysis of the hygroscopic water which had penetrated the sample.

The increase in the loss angle at elevated temperatures after irradiation of muscovite roasted at 600 and 800°C, and also of unroasted muscovite (especially noticeable at the lower frequency), is related to the increase in the electrical conductivity of the material as a result of the ionizing action of the radiation. This same fact explains the increased value of  $\epsilon$  for the irradiated sample of M-600 over that of the unirradiated one at a temperature above 120°C (Fig. 4b).

## Conclusions

1. The dielectric properties of unroasted muscovite are worsened after irradiation, which is due to the ionizing action of the gamma radiation.

2. Irradiation of muscovite roasted at 800°C leads to a sharp decrease in the loss angle over the temperature range from 20 to 100°C, and to an increase at higher temperatures.

3. In muscovite having a disintegrated structure, radiolysis of water is possible by irradiation, which can improve the dielectric characteristics of the mica.

In conclusion, the authors thank student A. V. Rand for taking part in the experiment.

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